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VARIATIONAL BOUNDS ON DARCY'S CONSTANT

James G. Berryman

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Lawrence Livermore National Laboratory

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James G. Berryman
Lawrence Livermore National Laboratory
P. O. Box 808, L-201
Livermore, CA 94550

Abstract

Prager's variational method of obtaining upper bounds on the fluid permeability (Darcy's constant) for slow flow through porous media is reexamined. By exploiting the freedom one has in choosing the trial stress distributions, several new results are derived. One result is a phase interchange relation for permeability; when the fluid-phase and particle-phase are interchanged for a fixed geometry, we find an upper bound on a linear combination of the complementary permeabilities. Another result is a proof of the monotone properties of the bounds. The optimal two-point bounds from this class of variational principles are evaluated numerically and compared to exact results of low density expansions for assemblages of spheres.

1. Introduction

Darcy's law [1] states that, when a viscous fluid moves slowly and steadily through a porous medium, the macroscopic flux (fluid volume crossing a unit cross-sectional area per unit time) is directly proportional to the pressure difference across the material and inversely proportional both to its thickness and to the viscosity of the fluid. The direction of flow is opposite to that of the positive pressure difference. The constant of proportionality in Darcy's law is called the fluid permeability or Darcy's constant. The law is modified slightly if gravity influences the fluid motion as it did in Darcy's original experiments; then the driving force is not simply the pressure difference but a linear combination of the forces due to gravity and pressure differential. For simplicity, gravitational effects will be neglected in the present discussion.

Darcy's law is a linear proportionality between the macroscopic flux and pressure gradient. This macroscopic relationship can be derived from the microscopic (Stokes) equations for slow flow of an incompressible fluid through a vessel of arbitrary shape with no slip between fluid and vessel at points of contact. (The terms "macroscopic" and "microscopic" are being used somewhat loosely here to distinguish between the two relevant length scales.) Derivations of Darcy's law using averaging methods have been given by Poreh and Elata [2] and by Neuman [3] while homogenization theory was used in the derivation by Keller [4]. The feature of this problem which makes it both especially interesting and especially difficult is the fact that, although only classical physics is involved at both the macroscopic and microscopic length scales, the macroscopic law of transport is not of the same form as the microscopic flow equations. The permeability is therefore a meaningful macroscopic concept without a direct microscopic analog. By contrast, the dielectric constant in Maxwell's equations is meaningful both at the microscopic level and at the macroscopic level for composites. Only when we consider much smaller (i.e. atomic) length scales does a similar change in the form of the relevant equations occur.

One early and fairly successful attempt to estimate the macroscopic permeability for flow through a random porous aggregate is the work of Brinkman [5]. By postulating a modified form of Darcy's law which might be expected to apply when the flow field is nonuniform, Brinkman was able to perform a single-site scattering calculation for flow around a spherical inclusion in a porous medium and then use this result to obtain an effective medium estimate of the permeability as a function of porosity. For very low densities of solid particles, Brinkman's approach gives the leading corrections to the Stokes drag on a spherical particle in the presence of other particles [6-8]. More recently some exact calculations of the permeability for periodic arrays of spheres have been performed by Zick and Homsy [9] and by Sangani and Acrivos [10]. All of these efforts are important benchmarks in the general theory of permeability but they fall short of helping us to estimate the permeability of an arbitrary porous random aggregate of particles.

Prager [11] introduced a very general variational principle for porous flow based on the concept of minimum energy dissipation. Upper bounds on permeability may be obtained from this minimum principle if certain statistical information concerning the topology of the pore space is available. Doi [12] introduced another variational principle for bounds on permeability but this approach does not have the flexibility inherent in Prager's method. Torquato [13] was the first to apply Prager's ideas to packings of penetrable and impenetrable spheres for which the required statistical correlation functions are known for large densities of particles. The present author repeated some of these calculations independently [14] and subsequently discovered that some different choices of trial stress distribution led to significantly different upper bounds on Darcy's constant [15]. The hope of obtaining realistic estimates of permeability when the required spatial correlation functions of an arbitrary porous specimen are known [16] remains unfulfilled at present but the progress which has been made will be reported here.

Section 2 presents the variational principle which leads to bounds on permeability. Section 3 explores the possible choices of trial stress distribution. Section 4 uses the freedom in choice of stochastic function in the trial stress distribution to derive a phase interchange relation for permeability. Section 5 discusses optimal upper bounds on permeability using only two-point spatial correlation functions which have been derived using the freedom in choice of deterministic function in the trial stress distribution. Section 6 illustrates the monotone properties of the bounds and summarizes our conclusions.

2. Minimum Energy Dissipation

In this section, we will present a complete derivation of the variational principle to be used in the remainder of the paper. The initial line of argument parallels that of the elastic problem as presented by Courant and Hilbert [17]. The conclusion of the derivation has been alluded to by Prager [11] and by Beran [18]; however, since some questions have been raised concerning a choice of normalization in the formulation [19,20], it will prove beneficial to include a full discussion here.

Equations for the Stokes flow of a viscous fluid through an arbitrary vessel of total volume Ω take the form

$$\Pi_{ij,j} = 0 \qquad \text{for } i, j = 1, 2, 3 \tag{2.1}$$

where the stress tensor is

$$\Pi_{ij} = -p\delta_{ij} + \sigma_{ij}, \tag{2.2}$$

the local fluid pressure is p, and the viscosity stress tensor is

$$\sigma_{ij} = \mu\,(v_{i,j} + v_{j,i}) \tag{2.3}$$

with μ the viscosity of the fluid. A subscript following a comma indicates a partial derivative. The local fluid velocity is v_i and if the fluid is assumed to be incompressible

$$v_{i,i} = 0. \tag{2.4}$$

The summation convention is assumed in both (2.1) and (2.4). The boundary conditions associated with (2.1) and (2.4) are the no slip condition

$$v_i = 0 \qquad \text{on} \qquad \Gamma_I \tag{2.5}$$

at interior points where the fluid touches the vessel and the stress matching condition

$$\Pi_{ij} = \overset{\circ}{\Pi}_{ij} \qquad \text{on} \qquad \Gamma_E \tag{2.6}$$

at points on the external boundary of the volume Ω. The fluid volume being considered is completely contained in the volume Ω_f whose surface is $\Gamma = \Gamma_I \cup \Gamma_E$. If the applied stress $\overset{\circ}{\Pi}_{ij}$ is not uniformly hydrostatic on Γ_E, then fluid will flow into Ω_f through parts of the surface Γ_E and out of Ω_f through other parts of Γ_E.

For simplicity, we will generally assume a slab geometry. The vessel is then a porous material lying between two parallel planes orthogonal to the z-axis. The applied stress takes the form of a uniform pressure p_- on one plane located at $z = -\Delta Z/2$ and $p_+ = p_- + \Delta P$ on the other at $z = \Delta Z/2$. If the thickness of the slab is ΔZ, then the pressure gradient is $\frac{\Delta P}{\Delta Z}$. The total volume Ω for the slab is infinite. The discussion which follows is phrased as if Ω is finite with the understanding that the limit $\Omega \to \infty$ will be taken at the end of the calculations.

Now we wish to reformulate (2.1)-(2.6) in terms of a variational principle. To do so, we introduce the quadratic form

$$Q(\sigma, \sigma) = \frac{1}{2\mu\Omega} \int_{\Omega_f} \sigma_{ij} \sigma_{ij} \; d^3 x \; . \tag{2.7}$$

Eq. (2.7) gives the rate of energy dissipation in the fluid per unit total volume [21]. Now if θ and τ are two symmetric, zero trace tensors and if θ is a viscosity stress tensor for some fluid velocity u_i, then

$$\theta_{ij} = \mu \left(u_{i,j} + u_{j,i}\right) \text{ in } \Omega_f \tag{2.8}$$

and

$$\begin{aligned} Q(\theta,\tau) &= \frac{1}{2\mu\Omega} \int_{\Omega_f} \theta_{ij} \; \tau_{ij} \; d^3x \\ &= -\frac{1}{\Omega} \int_{\Omega_f} u_i \; \tau_{ij,\,j} \, d^3x \; + \frac{1}{\Omega} \int_{\Gamma} u_i \tau_{ij} n_j ds. \end{aligned} \tag{2.9}$$

Eq. (2.9) is a Green identity which follows easily from an application of the divergence theorem; the infinitesimal surface element is ds and n_j is the j-th component of the unit outward normal.

Now define two comparison tensors θ° and τ° which satisfy the following conditions:

$$\theta^o_{ij} = \mu \left(u^o_{i,\,j} + u^o_{j,\,i}\right) \text{ in } \Omega_f \tag{2.10}$$

where u^o_i satisfies $u^o_{i,\,i} = 0$ (because of the zero trace property of θ°) and

$$u^\circ_i = 0 \qquad \text{on} \qquad \Gamma_I \;, \tag{2.11}$$

while

$$\overset{\circ}{\tau}_{ij,j} = \overset{\circ}{P}_{,i} \quad \text{in} \quad \Omega_f \tag{2.12}$$

and

$$\overset{\circ}{\tau}_{ij} = \overset{\circ}{\Pi}_{ij} + \overset{\circ}{p}\,\delta_{ij} \quad \text{on } \Gamma_E. \tag{2.13}$$

Consider symmetric, zero trace tensors θ and τ such that θ satisfies (2.10) and (2.11) for some velocity field u_i, not necessarily $\overset{\circ}{u}_i$, and that, like τ°, τ also satisfies equation (2.13) and another of the form (2.12) for some scalar ρ instead of p°. Then it follows from (2.9) that

$$Q\,(\theta - \theta^\circ, \tau - \tau^\circ) = 0 \tag{2.14}$$

for all such θ and τ. Thus, the tensors $\theta - \theta^\circ$ and $\tau - \tau^\circ$ are orthogonal with respect to Q.

With these definitions, we may now formulate two reciprocal [17] variational problems

$$Q\,(\theta - \tau^\circ, \theta - \tau^\circ) = \text{minimum} \tag{2.15}$$

and

$$Q\,(\tau - \theta^\circ, \tau - \theta^\circ) = \text{minimum}. \tag{2.16}$$

After eliminating known quantities, these two principles reduce to

$$Q(\theta, \theta) - \frac{2}{\Omega} \int_{\Gamma_E} u_i \overset{\circ}{\Pi}_{ij} n_j ds = \text{minimum} \tag{2.17}$$

and

$$Q(\tau,\tau) = \text{minimum}, \tag{2.18}$$

subject to the admissibility conditions (2.10) - (2.11) and (2.12) - (2.13) respectively. The absolute minimum for both (2.17) and (2.18) is achieved by the symmetric tensor $\sigma = \theta = \tau$ where σ satisfies (2.1) - (2.6). Furthermore, it is worth remarking that the admissibility conditions for one principle are the variational (or Euler) equations for the other and vice versa.

The first variational principle (2.17) is associated with the name of Helmholtz [11], [22]. This principle is difficult to apply in problems with random geometry because of the no slip condition (2.11) on the trial stress fields θ. A trivial but nevertheless admissible choice of trial velocity field is $u_i = 0$ everywhere. This choice places an upper bound of zero on the right hand side of (2.17). The general form of the minimum may be determined when $\theta = \sigma$ and $u_i = v_i$. Then, since it is straightforward to show that

$$Q(\sigma, \sigma) = \frac{1}{\Omega} \int_{\Gamma_E} v_i \overset{\circ}{\Pi}_{ij} n_j ds, \tag{2.19}$$

we find the minimum is given by

$$- \frac{1}{\Omega} \int_{\Gamma_E} v_i \overset{\circ}{\Pi}_{ij} n_j ds = \text{minimum}. \tag{2.20}$$

For the slab geometry, $\overset{\circ}{\Pi}_{ij} = - p_{\pm} \delta_{ij}$ on the two bounding planes. Thus,

$$\frac{1}{\Omega} \int_{\Gamma_E} v_i \overset{\circ}{\Pi}_{ij} n_j ds = \frac{-\Delta p}{\Omega} \int_{\Gamma_+} v_i n_i ds \tag{2.21}$$

where the surface Γ_+ is the plane with pressure p_+. We have used fluid incompressibility in simplifying (2.21); the volume of fluid flowing into the volume Ω_f across one plane must be matched by the volume of fluid flowing out of Ω_f across the other plane. Furthermore, the macroscopic flow U_z is defined by

$$U_z = \int_{\Gamma_+} v_i n_i ds/A \qquad (2.22)$$

where, for an isotropic porous material with porosity (void volume fraction) ϕ, A is total area of a cross section of the porous medium and the volumes are given by $\Omega = A\Delta z$ and $\Omega_f = \phi\Omega$. Thus, in terms of macroscopic quantities, (2.20) becomes

$$\frac{\Delta P}{\Delta Z} U_z = \text{minimum}. \qquad (2.23)$$

Now Darcy's law states that

$$U_z = -\frac{k}{\mu}\frac{\Delta P}{\Delta Z} \qquad (2.24)$$

where k is the permeability so (2.23) becomes

$$-\frac{k}{\mu}\left(\frac{\Delta P}{\Delta Z}\right)^2 = \text{minimum} \qquad (2.25)$$

for the variational problem (2.17). The zero upper bound on (2.24) implies a zero lower bound on k.

The minimum for (2.18) follows from the preceding arguments by taking $\tau = \sigma$. Then, we find

$$Q(\tau, \tau) \geq Q(\sigma, \sigma) = \frac{k}{\mu} \left(\frac{\Delta P}{\Delta Z}\right)^2. \tag{2.26}$$

Thus, the variational principle (2.18) may be rephrased as the statement that, of all symmetric, zero trace tensors τ satisfying the admissibility conditions (2.12) and (2.13), the unique choice $\tau = \sigma$ where σ satisfies (2.1) - (2.6) gives the minimum energy dissipation in the fluid. This principle is the fluid-dynamical version of Castigliano's principle for the equilibrium of an isotropic elastic body while the principle (2.17) is the fluid version of the principle of minimum potential energy in elasticity. The problem (2.18) is preferred for viscous flow because a large class of trial stress distributions is easily constructed as we will show in the next section.

To complete the formulation of the variational principle for Darcy's constant, some method of introducing information about the random geometry of the pore space must be provided. Following Prager [11], we introduce the stochastic function

$$g(\vec{x}) = \begin{cases} 1 \text{ if } \vec{x} \in \Omega_f \\ 0 \text{ if } \vec{x} \notin \Omega_f \end{cases} \tag{2.27}$$

and the volume average

$$\langle\cdot\rangle = \frac{1}{\Omega} \int_\Omega \cdot \quad d^3x \quad . \tag{2.28}$$

Then, the quadratic form Q becomes

$$Q(\tau, \tau) = \frac{1}{2\mu} \langle g \; \tau_{ij}\tau_{ij} \rangle \; . \tag{2.29}$$

Comparing (2.26) and (2.29), we see that it would be advantageous to express the pressure gradient in terms of the trial stress. Such an expression may be obtained using the admissibility conditions (2.12) and (2.13). First, define the macroscopic pressure gradient

$$G_i = \delta_{i3}\frac{\Delta P}{\Delta Z}. \tag{2.30}$$

Then, consider

$$\begin{aligned}\langle g\tau_{ij,j}\rangle &= \frac{1}{\Omega}\int_{\Omega_f}\tau_{ij,j}d^3x \\ &= \frac{1}{\Omega}\int_{\Gamma_E} n_i\rho ds + \frac{1}{\Omega}\int_{\Gamma_I} n_i\rho ds.\end{aligned} \tag{2.31}$$

Since the trial stress distribution satisfies (2.13) and since the trial pressure field $\rho = p^0$ on Γ_E, we have

$$\frac{1}{\Omega}\int_{\Gamma_E} n_i\rho ds = \delta_{i3}\frac{(P_+ - P_-)}{\Omega}\int_{\Gamma_E} ds = \phi G_i. \tag{2.32}$$

Furthermore, since $\tau_{ij} = 0$ on Γ_E, the divergence theorem gives

$$\frac{1}{\Omega}\int_{\Omega_f}\tau_{ij,j}d^3x = \frac{1}{\Omega}\int_{\Gamma_I}\tau_{ij}n_j ds, \tag{2.33}$$

so the normalization condition may be expressed as

$$\frac{1}{\Omega}\int_{\Gamma_I}(\tau_{ij} - \rho\delta_{ij})\,n_j ds = \phi G_i. \tag{2.34}$$

Eq. (2.34) is the correct normalization on a general trial stress τ. This normalization is the same one used by Weissberg and Prager [19] but it differs from the one used originally by Prager [11]. Ramifications of the switch from the erroneous normalization to the correct one have been discussed by Berryman and Milton [20]. Using (2.26), (2.29), (2.30), and (2.34), the variational principle may now be written as

$$k \leq \frac{1}{2}\langle g\tau_{ij}\tau_{ij}\rangle / G_k G_k. \tag{2.35}$$

As a final check on the variational principle, we perform a first variation of (2.35). As formulated using (2.34), the variation of the denominator vanishes identically. The variation of the numerator is

$$\delta\langle g\tau_{ij}\tau_{ij}\rangle = 2\langle g\tau_{ij}\,\delta\tau_{ij}\rangle. \tag{2.36}$$

If $\tau_{ij} = \mu\,(w_{i,j} + w_{j,i})$ for some vector w_i, then it follows easily that

$$< g\tau_{ij}\delta\tau_{ij} > = \frac{2\mu}{\Omega} \int_{\Gamma} w_i(\delta\tau_{ij} - \delta\rho\,\delta_{ij})\,n_j ds + \frac{2\mu}{\Omega} \int_{\Omega_f} w_{i,i}\,\delta\rho\,d^3x \tag{2.37}$$

The zero trace property of τ_{ij} guarantees that $w_{i,i} = 0$ in Ω_f so, if we define $\Pi_{ij} = -\rho\delta_{ij} + \tau_{ij}$, then

$$\delta < g\tau_{ij}\tau_{ij} > = \frac{4\mu}{\Omega} \int_{\Gamma_I} w_i\,\delta\Pi_{ij}n_j ds + \frac{4\mu}{\Omega} \int_{\Gamma_E} w_i\,\delta\mathring{\Pi}_{ij}\,n_j ds \quad . \tag{2.38}$$

The second surface integral in (2.38) vanishes because $\mathring{\Pi}_{ij}$ is specified on the external boundary. The variation (2.34) therefore vanishes if $w_i = 0$ on Γ_I since the variations $\delta\Pi_{ij}$ may be arbitrary there. Thus, the ratio in (2.35) is stationary if the admissible trial stress τ satisfying (2.12) and (2.13) also satisfies (2.10) and (2.11). A second variation of (2.35) produces a positive result; hence, the stationary point is in fact a minimum.

This calculation concludes the formulation of the variational principle. The next section discusses methods of choosing useful trial stress distributions.

3. Trial Stress Distributions

Trial stress distributions τ for the variational bound (2.35) on Darcy's constant must satisfy the following admissibility conditions:

$$\tau_{ij} = \tau_{ji}, \tag{3.1}$$

$$\tau_{kk} = 0, \tag{3.2}$$

$$\tau_{ij,j} = \rho_{,i} \text{ in } \Omega_f, \tag{3.3}$$

$$\tau_{ij} = \overset{\circ}{\Pi}_{ij} + \overset{\circ}{p}\delta_{ij} \text{ on } \Gamma_E, \tag{3.4}$$

and

$$\frac{1}{\Omega}\int_{\Gamma_I}(\tau_{ij} - \rho\delta_{ij})n_j ds = \phi\frac{\Delta P}{\Delta Z}\delta_{i3}. \tag{3.5}$$

We may replace the admissibility condition (3.3) by the equivalent condition

$$\epsilon_{mik}\tau_{ij,jk} = 0. \tag{3.6}$$

In (3.6), ϵ_{mik} is the Levi-Civita symbol (defined to be zero if any two of the indices are equal and either +1 or -1 for even or odd permutations of 123) so (3.3) has been replaced by the statement that the curl of a gradient vanishes identically. The principal effect of (3.5) is to show that τ and ρ must be correlated with the stochastic function g and the unit outward normal vector n_i. However, the required correlation of g and τ is not a very stringent condition; many examples of suitable choices for τ could be listed including various functions and functionals of g.

The most serious difficulty with the application of the variational principle is incomplete knowledge of g. In computer experiments on flow through random aggregates, it is possible to have as much information as desired about the stochastic function. In any other circumstance of practical interest, we should assume from the outset that only limited knowledge of the statistical properties of the porous medium will be available and design our trial functions to use the information at hand. Prager [11] provided one solution to this problem by introducing the trial stress distribution

$$\tau_{ij}(\vec{x}) = \int T_{ij}(\vec{r})\, h(\vec{x} + \vec{r})\, d^3r, \qquad (3.7)$$

where T_{ij} is a deterministic tensor and h is some stochastic scalar and the integral is over all space. The deterministic part of this functional $T_{ij}(\vec{r})$ may then be required to satisfy

$$T_{ij} = T_{ji}, \qquad (3.8)$$

$$T_{kk} = 0, \qquad (3.9)$$

and

$$T_{ij,j} = \psi_{,i} \qquad (3.10)$$

for some deterministic scalar function ψ or equivalently

$$\epsilon_{mik}\, T_{ij,jk} = 0 \qquad (3.11)$$

in order to guarantee satisfaction of (3.1), (3.2), and (3.6). Prager made the particularly simple choice

$$h(\vec{x}) = g(\vec{x}) \qquad (3.12)$$

for the stochastic scalar; however, we will show in the following sections that other choices of h are both possible and preferable in some cases. Along with the choice (3.12), we make the assumption that g $(\vec{x})$ may be extended outside the volume Ω so that the integral in (3.7) may be evaluated. A periodic extension of g is conceptually simple but undesirable because of the long range order introduced this way. A superior choice of extension is to assume that other samples are drawn from the same ensemble as g and arranged to fill all space. This extension causes no conceptual or practical difficulties as we will see. The boundary condition (3,4) on Γ_E will be satisfied by (3.7) if we assume the microstructure of the porous medium is much smaller than the macroscopic scale of the slab.

Using linearity with respect to the applied pressure gradient and the symmetric nature of T from (3.8) while assuming that the medium is isotropic, Prager obtained the general form of the deterministic function. In general, this result becomes

$$\begin{aligned} T_{ij}(\vec{r}) = \ &\{\alpha(r)\,[r_j G_i + r_i G_j] \\ &+ \beta(r) G_k r_k r_i r_j + \gamma(r) G_k r_k \delta_{ij}\}. \end{aligned} \qquad (3.13)$$

The scalar functions α, β, and γ depend only on the magnitude $r = |\vec{r}|$. The tensor (3.13) will satisfy (3.9) if

$$\gamma(r) = -\frac{1}{3}\,[2\alpha(r) + r^2\,\beta(r)]. \qquad (3.14)$$

T will also satisfy (3.11) if, for $i = k$,

$$0 = T_{ij,jk} - T_{kj,ji} \tag{3.15}$$

$$= (\alpha'' + \frac{4\alpha'}{r} - 5\beta - r\beta')(G_i r_k - G_k r_i).$$

Thus, we find that

$$\frac{d}{dr}[r^5\beta(r)] = \frac{d}{dr}[r^4 \frac{d\alpha(r)}{dr}] \tag{3.16}$$

or equivalently that

$$r^5\beta(r) = r^4 \frac{d\alpha(r)}{dr} + \text{const.} \tag{3.17}$$

Except for the constant appearing in (3.17), β and γ are completely determined by (3.17) and (3.14) once a functional form for α has been chosen. Several choices for the function α will be discussed in Section 5.

The constant in (3.17) is fixed by the normalization condition (3.5). To see this, substitute (3.13) into (3.10) and note [20] that a solution for ψ is

$$\psi(\vec{r}) = G_k r_k [r \frac{d\alpha(r)}{dr} + \frac{10}{3}\alpha(r) - \frac{1}{3} r^2 \beta(r)]. \tag{3.18}$$

Furthermore, the trial pressure field p is given by

$$p(\vec{x}) = \int \psi(\vec{r})\, h(\vec{x} + \vec{r})\, d^3r. \tag{3.19}$$

After some simplification, we find [20] that (3.5) becomes

$$\int d^3r[T_{ij}(\vec{r}) - \psi(\vec{r})\delta_{ij}] \frac{r_j}{r} \frac{dS_2(r)}{dr} = \phi G_i \tag{3.20}$$

where $S_2(r)$ is the two-point (auto-) correlation function of $g(\vec{x})$ satisfying $S_2(0) = \phi$ and $S_2(\infty) = \phi^2$. Then Eq. (3.20) reduces to the statement that

$$\frac{1}{3}\int d^3r\, r^2[r\beta(r) - \frac{d\alpha(r)}{dr}] \frac{dS_2(r)}{dr} = -\frac{4\pi}{3}\phi(1-\phi)\,\text{const} = \phi \tag{3.21}$$

using (3.17). So the constant in (3.17) is just

$$\text{const} = -\frac{3}{4\pi(1-\phi)}. \tag{3.22}$$

4. Phase Interchange Relation

In this section, we will use the flexibility in choice of stochastic function in (3.7) to derive a phase interchange relation for permeability. By a phase interchange relation, we mean any equality or inequality which applies to a sum or product of the effective constants when the geometry of the material is held fixed but the roles of the constituents are interchanged. Keller [23] derived such a phase interchange equality for the products of conductivities of two-phase composites in two dimensions. Keller's results were generalized to an inequality for products of conductivities of two-phase composites in three dimensions by Schulgasser [24]. We will derive an inequality for a linear combination of the permeabilities for a porous medium and its complement.

The complementary medium has the same geometrical structure as the original medium but the locations of solid and void are interchanged. The mathematical consequence of this interchange is that the stochastic function $\bar{g}$ for the complement is

$$\bar{g}(\vec{x}) = 1 - g(\vec{x}) \tag{4.1}$$

if the stochastic function g for the original medium is given by (2.27). Using the bar to distinguish properties of the complementary medium, we find easily that the porosity of the complement is

$$\bar{\phi} = 1 - \phi \tag{4.2}$$

and we define the complementary permeability to be $\bar{k}$.

Now it will prove to be illuminating to consider trial stress distributions of the form (3.7) for the original porous medium with either of two choices for h

$$h_1(\vec{x}) = g(\vec{x}) \tag{4.3}$$

or

$$h_2(\vec{x}) = \bar{g}(\vec{x}) \tag{4.4}$$

and for T_{ij} we choose the (so-called) constant trial function given by (3.17) with $\alpha = 0$. Making these substitutions into (2.31), we find that the results depend on certain spatial correlation functions of the form

$$C_2^{(m)}(\vec{r}) = \langle g(\vec{x})h_m(\vec{x}+\vec{r})\rangle \tag{4.5}$$

and

$$C_3^{(m)}(\vec{r},\vec{s}) = \langle g(\vec{x})h_m(\vec{x}+\vec{r})h_m(\vec{x}+\vec{s})\rangle . \tag{4.6}$$

The two variational bounds associated with the (4.3) and (4.4) are then

$$k \leq \iint T_{ij}(\vec{r})T_{ij}(\vec{s})C_3^{(m)}(\vec{r},\vec{s})d^3r d^3s/2G_k G_k \tag{4.7}$$

for m=1 and 2.

Two important identities follow for (4.3) - (4.6):

$$C_{2,j}^{(2)}(\vec{r}) = - C_{2,j}^{(1)}(\vec{r}) \tag{4.8}$$

and

$$C_3^{(2)}(\vec{r},\vec{s}) = \phi - C_2^{(1)}(\vec{r}) - C_2^{(1)}(\vec{s}) + C_3^{(1)}(\vec{r},\vec{s}). \qquad (4.9)$$

If we substitute (4.9) into the numerator of (4.7) for $m = 2$, we find that the first three terms on the right hand side of (4.9) do not contribute to the double integral if T_{ij} is of the form (3.13) since it is easily shown that

$$\int T_{ij}(\vec{r})d^3r = 0 \quad . \qquad (4.10)$$

The physical significance of (4.10) is that the mean stress deviations must vanish $<\tau_{ij}> = 0$ since the system as a whole is not being sheared. Thus, the numerators of both bounds are equal and the bounds for either choice (4.3) or (4.4) are the same. This result shows that the bounds depend most strongly on the arrangement of the internal surface which is identical for the sample and its complement.

Now consider the related spatial correlation functions

$$\bar{C}_2^{(m)}(\vec{r}) = <\bar{g}(\vec{x})h_m(\vec{x}+\vec{r})> \qquad (4.11)$$

and

$$\bar{C}_3^{(m)}(\vec{r},\vec{s}) = <\bar{g}(\vec{x})h_m(\vec{x}+\vec{r})h_m(\vec{x}+\vec{s})>. \qquad (4.12)$$

Then it is straightforward to show that

$$\bar{C}_{2,j}^{(m)}(\vec{r}) = -C_{2,j}^{(m)}(\vec{r}) \qquad (4.13)$$

and

$$\overline{C}_3^{(m)}(\vec{r},\vec{s}) + C_3^{(m)}(\vec{r},\vec{s}) = \langle h_m(\vec{x}+\vec{r})h_m(\vec{x}+\vec{s})\rangle .$$

$$\equiv S_2^{(m)}(|\vec{r}-\vec{s}|) \quad . \tag{4.14}$$

Using (4.13), the phase-interchanged version of (4.7) becomes

$$\overline{k} \leq \left(\frac{\overline{\phi}}{\phi}\right)^2 \int\int T_{ij}(\vec{r})T_{ij}(\vec{s})\overline{C}_3(\vec{r},\vec{s})d^3r\ d^3s/2G_kG_k \ , \tag{4.15}$$

where we have accounted for the difference in normalization constant by the factor preceding the integral. Multiplying (4.15) by $(\phi/\overline{\phi})^2$, adding the result to(4.7) and using (4.14), we have

$$k + \left(\frac{\phi}{\overline{\phi}}\right)^2 \overline{k} \leq \phi^2 \int\int T_{ij}(\vec{r})\ T_{ij}(\vec{s})\ S_2^{(m)}(|\vec{r}-\vec{s}|)d^3r\ d^3s/2G_kG_k . \tag{4.16}$$

The inequality (4.16) is a phase interchange relation for this linear combination of the permeability k and its complementary value $\overline{k}$.

To begin analyzing (4.16), first notice that this upper bound depends only on two-point spatial correlation functions. Next notice that, since both contributions to the left hand side of (4.16) are strictly positive, we may eliminate either term and still have a valid upper bound on the remaining term. Thus, in (4.7) $C_3^{(m)}$ may be replaced by $S_2^{(m)}$ while maintaining the validity of the inequality. Such a replacement has some advantages for practical applications because two-point spatial correlation functions are more easily measured or computed than three-point correlations.

The two-point upper bound in (4.16) is not a new result. Prager [11] showed that, since $g \leq 1$, the factor of g in the numerator of (2.35) can be replaced by unity and still have a valid bound on Darcy's constant. The result is

$$k \leq \langle \tau_{ij} \tau_{ij} \rangle / 2 G_k G_k \ , \tag{4.17}$$

which has the same right hand side as (4.16). The new result (4.16) therefore shows that the bound (4.17) must be a poor estimate of k if $\bar{k}$ is finite. If ϕ is large, then it will often be the case that there are no connecting paths through the particle-phase; then the permeability $\bar{k}$ for the interchanged problem will vanish identically. Thus, it is possible for (4.17) to provide a good estimate of k when $\phi \to 1$, as has been observed in numerical calculations [13-15, 20]. Similarly, if ϕ is small, then it is possible that no connecting paths through the void-phase exist; then the permeability k will vanish identically. But (4.16) shows that the right hand side of (4.17) is bounded away from zero even when $k = 0$ because $\bar{k}$ will be finite (and large) in this limit. This argument shows why the two-point bounds (4.17) (which are evaluated numerically in the next section) provide such bad estimates of k at low porosities.

5. Two-Point Bounds

Attention will now be restricted to two-point bounds to provide some elaboration of the results on the phase interchange relation (4.16). It has been shown elsewhere [20] that the best possible two-point bound obtainable using the trial stress distribution (3.7) with (3.12) has the form

$$k \le \frac{2}{3} \int_0^\infty dr\, r[S_2(r) - \phi^2]/(1-\phi)^2 \ , \tag{5.1}$$

where $S_2 = S_2^{(1)}$ defined in (4.14). The relation (5.1) was found by noting that, if the deterministic function T_{ij} is allowed to vary over the entire class of admissible tensors, the smallest value of $1/2\langle\tau_{ij}\tau_{ij}\rangle$ is the right hand side of (5.1).

To permit the evaluation of (5.1), the discussion must also be limited to a particular kind of random geometry for which the various two-point spatial correlation functions are known. Until such data become available for real materials [16], we are essentially limited to two types of model materials: (1) random packings of impenetrable spheres and (2) random assemblages of penetrable spheres. Two-point bounds for impenetrable-sphere packs have been discussed in [14]. For the present discussion, we restrict our analysis to the penetrable sphere model.

The penetrable sphere model assumes that particle centers are distributed randomly in the volume Ω and that each center is surrounded by a sphere of particle material. In the simplest version of this model, all these spheres have the same radius R. If the density of particle centers is great enough and the sphere radius large enough, at least some and in general many of these spheres will overlap. This model has the distinct advantage that analytical results are known for all the spatial correlation functions of interest [12,15,25-27].

Since the particle centers are uncorrelated, it is not difficult to show [25-27] that the general result for an n-point void correlation function is

$$S_n(\vec{x}_1,\ldots,\vec{x}_{n-1}) \equiv \langle g(\vec{x})g(\vec{x}+\vec{x}_1)\ldots g(\vec{x}+\vec{x}_{n-1})\rangle = \exp(-\rho V_n) \tag{5.2}$$

where ρ is the number density of spheres and V_n is the union volume of n spheres with the fixed radius R and centers at the vertices $\vec{x}_1,\ldots, \vec{x}_{n-1}$. The union volume for one sphere is just

$$V_1 = \frac{4\pi}{3}R^3, \tag{5.3}$$

so the porosity is given by

$$\phi = S_1 = \exp(-\rho \frac{4\pi}{3}R^3). \tag{5.4}$$

For two spheres, the union volume is found to be

$$V_2(\zeta R)/R^3 = \begin{cases} \frac{4\pi}{3}\,(1+\frac{3}{4}\zeta-\frac{\zeta^3}{16}) & \text{for } \zeta\leq 2 \\ \frac{8\pi}{3} & \text{for } \zeta\geq 2 \end{cases} \tag{5.5}$$

and S_2 follows from (5.2).

The results of computations using the penetrable sphere model to provide the data needed for the two-point correlation functions are summarized in Table I and Figure 1. For comparison, the analytical result of Weissberg and Prager [19] that

$$k \leq -\frac{2}{9}\,R^2\ \frac{\phi}{\ln\phi} \tag{5.6}$$

is also shown in Figure 1. The inequality in (5.6) was derived for the penetrable sphere model using the same variational principle discussed in Section 2, but a different trial function which takes advantage of the symmetries inherent in assemblages of penetrable spheres.

Also shown is the low density expansion for the permeability of an assemblage of hard spheres [6-8]

$$k = \frac{2}{9}\ \frac{R^2}{\eta}[1 + \frac{3}{\sqrt{2}}\eta^{1/2} + \frac{135}{64}\eta\ln\eta + 16.5\eta + \ldots]^{-1}, \qquad (5.7)$$

where the solid volume fraction is $\eta = 1-\phi$.

Table I and Figure 1 show that the result of Weissberg and Prager is somewhat superior to the two-point bound of Prager for the penetrable sphere model. However, two distinctions should be stressed: (1) The variational bound (5.1) is valid for any isotropic porous solid whereas (5.6) is valid only for the penetrable sphere model. (2) Because the derivation of (5.6) differs significantly from that of (5.1), it cannot be said that (5.6) is a "two-point bound". Indeed the derivation of (5.6) avoids the introduction of the spatial correlation functions altogether. Unfortunately, the trick which leads to (5.6) appears to work only for the penetrable sphere model, so (5.1) appears to be the best general bound on permeability using the limited information in the two-point correlation functions.

6. Discussion

As a final example of the versatility of the variational principle (2.35), consider two porous media with stochastic functions f and g whose porosities are respectively

$$\langle f\rangle = \phi_f \quad \text{and} \quad \langle g\rangle = \phi_g \,. \tag{6.1}$$

Then, if we define the right hand side of (2.35) to be $k_g(\tau)$, we have

$$k \leq k_g(\tau) = \langle g\tau_{ij}\tau_{ij}\rangle / 2G_kG_k \,. \tag{6.2}$$

Now suppose that the stochastic functions of these two materials are related by the inequality

$$f(\vec{x}) \geq g(\vec{x}) \text{ for all } \vec{x}, \tag{6.3}$$

so the material characterized by f is more porous than the one for g; in particular, the material with stochastic function g is obtained by adding more solid material to the other one without rearranging the original material. Then, it is clearly true that

$$\langle g\tau_{ij}\tau_{ij}\rangle \leq \langle f\tau_{ij}\tau_{ij}\rangle \tag{6.4}$$

for any given trial stress distribution τ. Substituting (6.4) into (6.2) and defining $k_f(\tau)$ as the right hand side of (2.35) with g replaced by f, we find

$$k \leq k_g(\tau) \leq k_f(\tau) \tag{6.5}$$

where k is the actual permeability for the material characterized by g.

The inequalities in (6.5) are true for any fixed τ. In particular, if $\tau = \sigma_f$ where σ_f minimizes k_f, then

$$k \leq k_g\ (\sigma_f) \leq k_f(\sigma_f). \tag{6.6}$$

Similarly, if σ_g minimizes k_g, then

$$k = k_g\ (\sigma_g) \leq k_f\ (\sigma_g). \tag{6.7}$$

Thus, it follows from (6.6) and (6.7) that

$$k = k_g\ (\sigma_g) \leq k_g\ (\sigma_f) \leq k_f\ (\sigma_f). \tag{6.8}$$

This argument shows that, if one porous material differs from a more porous one only by the addition of solid material (with no rearrangement of the other material), then--as one might expect intuitively--the permeability of the more porous material is always greater than or equal to that of the less porous one.

The inequalities in (6.5) have other practical consequences. If we use the same trial function τ for two problems with f and g related by (6.3), then the bound obtained for the more porous material will be a valid bound for the less porous one. This relationship is observed to be satisfied by the examples in Table I for the penetrable sphere model. Specific representations of the penetrable sphere model can satisfy (6.3) very easily by choosing a particular set of sphere centers and letting the radii of the overlapping spheres satisfy $R_f < R_g$ for the two cases. Thus, (6.5) is a useful check on our numerical integration method and also guarantees that the curves in Figure 1 will decrease monotonically as observed.

In conclusion, the main new result of the present work is the observation that the stochastic function h appearing in the trial stress distribution (3.8) can take a variety of forms. We have used the freedom in choosing both the deterministic function T_{ij} and the stochastic function h in (3.8) to obtain several new results. A phase interchange relation for a linear combination of the permeability and its complementary value was obtained in (4.16). A proof of the monotone properties of the bounds was given in (6.8). Numerical comparisons of bounds on permeability were made for the penetrable sphere model when the limited geometrical information contained in the two-point spatial correlation functions is available. Future developments in the theory will require knowledge of three-point and possibly higher order spatial correlation functions.

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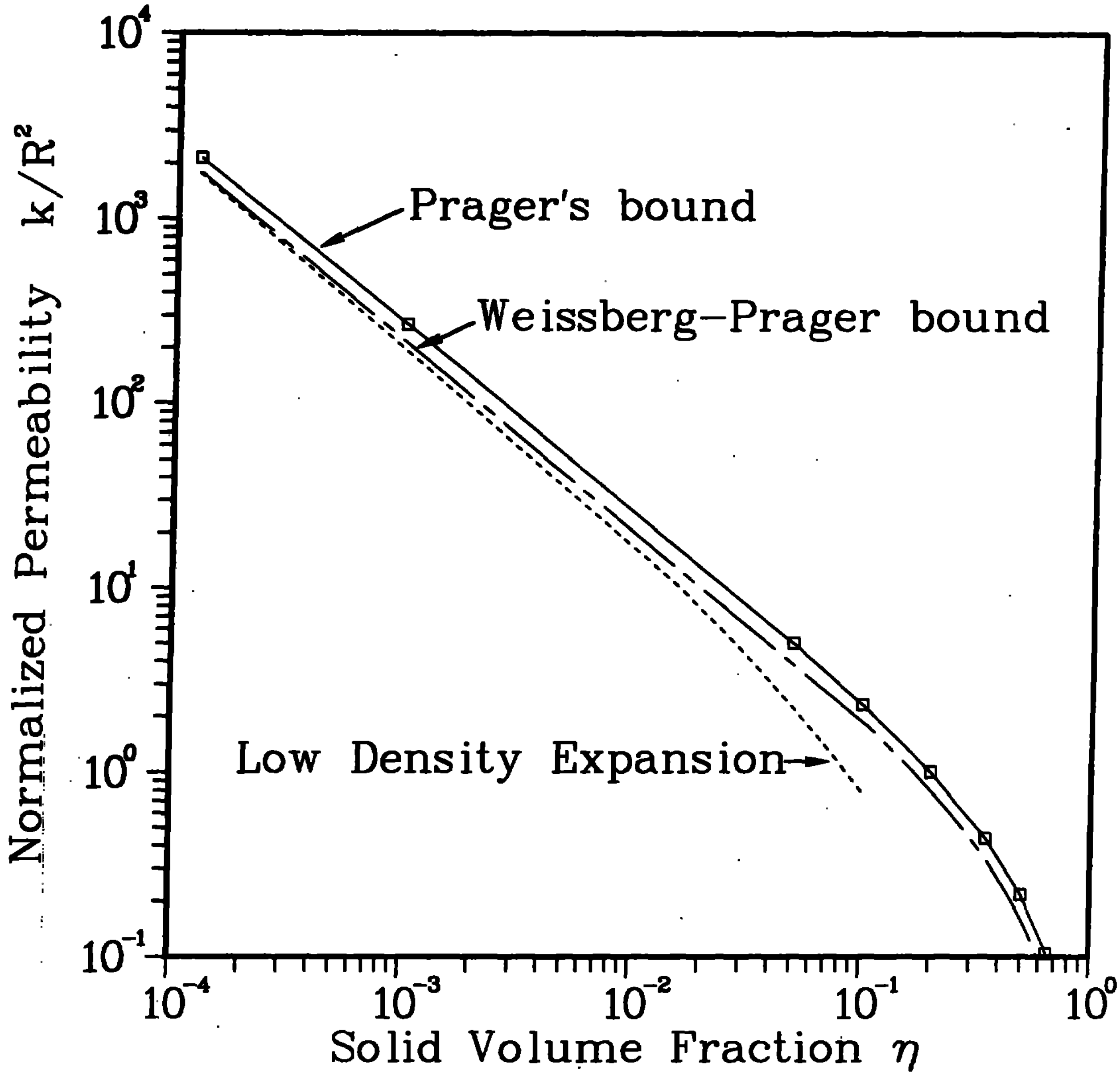
η	k_{WP}/R^2	k_p/R^2
0.1	1.898E+00	2.329E+00
0.2	7.967E-01	1.001E+00
0.3	4.361E-01	5.611E-01
0.4	2.610E-01	3.443E-01
0.5	1.603E-01	2.170E-01
0.6	9.701E-02	1.348E-01
0.7	5.537E-02	7.885E-02
0.8	2.761E-02	4.009E-02
0.9	9.651E-03	1.395E-02

Table I. Values of the upper bounds on permeability due to Weissberg and Prager (k_{WP}) and due to Prager (k_p). The corresponding formulas in the text are respectively (5.6) and (5.1).

Figure Caption

Figure 1. Comparison of Prager's two-point bound (boxes and solid line) to Weissberg and Prager's analytical bound (long-dash/short-dash line) for penetrable spheres and to the low density expansion for hard spheres (dashed line) as the porosity ϕ or solid volume fraction $\eta = 1-\phi$ varies. The corresponding equations in the text are respectively (5.1), (5.6), and (5.7). Some numerical values are also listed in Table 1.



Prager's bound
Weissberg–Prager bound
Low Density Expansion
Normalized Permeability k/R²
Solid Volume Fraction η
10⁴
10³
10²
10¹
10⁰
10⁻¹
10⁻⁴
10⁻³
10⁻²
10⁻¹
10⁰

FIGURE 1